

**CHARACTERIZATION AND PERFORMANCE EVALUATION OF AN HPXe DETECTOR
FOR NUCLEAR EXPLOSION MONITORING APPLICATIONS**

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ABSTRACT

Expanding missions in nuclear explosion monitoring (NEM) and nuclear security have highlighted the need for high-resolution ambient-temperature gamma detectors that can provide radionuclide-specific monitoring under demanding field conditions. Recent improvements in high-pressure xenon (HPXe) detectors indicate that this technology has potential to provide rugged, large volume ambient temperature gamma detectors with adequate resolution for radionuclide analysis to meet needs in several mission areas.

The purpose of this Phase I study was to evaluate the feasibility of HPXe-based monitoring systems for meeting required detection sensitivity limits for ¹⁴⁰Ba for specified NEM sampling and counting conditions. An HPXe detector was selected and characterized for the NEM application. A series of experimental measurements with a custom NIST-traceable 9-radionuclide source were conducted to define the energy, efficiency and resolution performance of the detector, and to compare the performance with sodium iodide and germanium detectors.

Monte Carlo (MCNP) simulation was used to select optimum air filter geometries (concentric cylinder), to examine efficiency improvements for aluminum vs. steel detector wall material (aluminum ~50% more efficient), and to estimate optimum shield dimensions for an HPXe based nuclear explosion monitor.

MCNP modeling was also used to estimate the detection sensitivity of the HPXe detector for the nuclear explosion fission product indicator, ¹⁴⁰Ba. Background spectra for the HPXe detector were calculated with MCNP by using input activity levels as measured in routine NEM runs at Pacific Northwest National Laboratory (PNNL). Analysis of the composite spectra indicates that the required detection sensitivity for ¹⁴⁰Ba can likely be met using the 537 keV gamma peak in the composite spectrum of the HPXe detector. Based on the Phase I project results, a design concept was defined for a NEM Prototype to be developed in the Phase II program.

HPXe appears to be a candidate for applications requiring simple, high reliability ambient temperature radionuclide measurement systems with slightly less energy resolution capability than that of cooled germanium systems. Unfulfilled applications exist now for these performance capabilities in nuclear non-proliferation, homeland security, nuclear site cleanup, nuclear power, and portable instruments for health physics needs.

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OBJECTIVES

The overall purpose of this R&D project is to evaluate potential performance benefits of ambient-temperature HPXe detectors for applications in the NEM program. The project is focused on matching the capabilities of HPXe detector systems with both the measurement requirements and operational considerations for specific NEM applications.

Specific objectives of Phase I project were as follows:

1. Identify a promising HPXe detector for NEM application; measure the basic detector parameters to provide data for calculations of performance and detection sensitivity limits for specific NEM applications.
2. Develop and verify an MCNP model for HPXe-based systems that can provide quantitative evaluation of the performance for HPXe-based systems and optimization of designs for specific NEM applications.
3. Define a conceptual design for an HPXe-based NEM prototype system for field testing.

RESEARCH ACCOMPLISHED

Background

Radioisotope identification for NEM by gamma spectroscopy requires extremely reliable high-resolution and high-efficiency gamma detectors. Although high purity germanium (HPGe) detectors now represent the energy resolution performance standard for laboratory gamma spectroscopy, their inherent reliability issues and requirement for cryogenic cooling present practical operational problems in applications for field or remote nuclear security monitoring. The final summary slide of a recent seminar (Aiken, 2007) with participation by eight federal government agencies with nuclear security missions listed the first priority item needed for improved nuclear detection and response capability as: "New detector materials to provide high resolution ambient temperature operation."

A brief summary of the current status of germanium detectors and of candidates for non-cooled detectors for radionuclide specific nuclear security applications is shown in Table 1. The two alternate technologies that appear to hold the most immediate potential for higher resolution ambient temperature detectors for nuclear security are La-halide scintillation crystals and high-pressure xenon chambers.

Table 1: Candidate Detector Technologies for Nuclear Explosion Monitoring

Detector Type	Resolution at 662 keV	Active Volume	Stability	Reliability	Practical Limitations
HPGe	0.2-0.5 %	moderate	Excellent	Moderate	Need for cryogenic cooling & vacuum, mechanical ruggedness, size limit, cost
HPXe	1.7-2.4 %	Excellent	Excellent	Excellent	Low density, less high energy efficiency, cost, evolving technology
CdZnTe	2-3 %	Poor	Poor	Moderate	Very small size, poor long-term stability, cost, evolving technology
La-Halides	2.5-4 %	Moderate	Moderate	Unknown	Temperature dependent output, natural radioactivity, cost, evolving technology
NaI	7-9 %	Excellent	Moderate	Excellent	Insufficient resolution, drift, crystal fracture

The goals of this R&D project are to evaluate HPXe detector performance as applied to improvements in nuclear explosion monitoring, and to develop and test a prototype system that will allow prompt field evaluation of promising NEM applications. This paper describes the Phase I effort consisting of selection and characterization of HPXe detectors for NEM applications, and of conceptual design of a NEM system prototype unit to be developed in the Phase II project.

The first activity of the Phase I project was selection of an HPXe detector suited for NEM applications and characterization of the detector performance by experimental measurements and MCNP5 simulations. Detection sensitivity for an HPXe-based NEM system was then estimated by combining the performance characterization data with the reported background data from field operation of radionuclide aerosol sampler analyzer (RASA) systems. Design concepts for the NEM prototype unit to be built in the Phase II effort were then defined based on the Phase I results.

Detector Selection and Characterization

Detector Selection - The primary performance benchmarks for the HPXe-based NEM are the specifications of the National Aeronautics and Space Administration solicitation and of the International Monitoring System (IMS) requirements for certification. As reported in the literature, the IMS required detection sensitivity specification is met by both the PNNL RASA system with a 90% HPGe detector (Miley, 1998) and by the EML AUTORAMP system with a 30% HPGe detector (Lanner, 1977). The active volume of a germanium detector of 90% relative efficiency is about 350 cc.

The density of germanium (5.3 g/cc) is about 13 times greater than that of xenon as compressed to 0.4 g/cc in the HPXe detector. However, the photopeak efficiency per unit volume is much closer for the two materials, since photoelectric cross section in the energy range below 600 keV is proportional to between Z^4 and Z^5 . Thus, the photoelectric cross section of xenon ($Z=54$) is about 8–14 times greater than for germanium ($Z=32$) over the 100–600 keV energy range of interest for detection and quantification of ^{140}Ba .

In view of the atomic number and density differences between HPXe detectors and germanium detectors, a Frisch-grid HPXe detector of active dimensions 11.4 cm diameter by 20.3 cm length (4.5 in. diameter X 8 in. length) and 2.4% full width at half maximum (FWHM) resolution at 662 keV was selected for testing as a candidate to meet the IMS sensitivity spec. The test HPXe detector, shown in Figure 1, has an active volume of more than 1900 cc, or more than 5 times that of the 90% germanium detector. The increased diameter and length for this HPXe detector as compared to the 90% germanium detector also allows the use of a single filter of large area for NEM monitor applications.



Figure 1 HPXe Test Detector (Front) during Test Series at UC-Irvine

Detector Characterization - Authoritative energy and efficiency calibration data were obtained for the selected HPXe detector by obtaining measured spectra using a custom 9-radionuclide point source with NIST traceability.

All counting experiments were performed at the University of California -Irvine research reactor facility using commercial radionuclide sources and special sources prepared by reactor irradiations. The calibration parameter measurements were made with the point source positioned at 25 cm from the detector axis at the midpoint of the detector length.

The energy versus channel number measurements at ten energy points from 88 keV to 1836 keV were fitted with insignificant residual differences to a linear equation for energy versus channel number : $E = -7.596 + 1.013$. As expected, the energy response of the HPXe detector was very linear.

FWHM energy resolution measurements were also made for the HPXe detector at energies of the peaks of the 9-radionuclide calibration source. Energy resolution was calculated using the Gaussian fitting routine of the Genie 2000™ Gamma Acquisition and Analysis software. The resolution equation with coefficients fitted to the measured resolution of the HPXe detector (2.4% FWHM at 662 keV) is shown in Figure 2. An MCNP calculated case for a detector with the same dimensions as the test detector, but with 2.0% resolution at 662 keV is also shown.

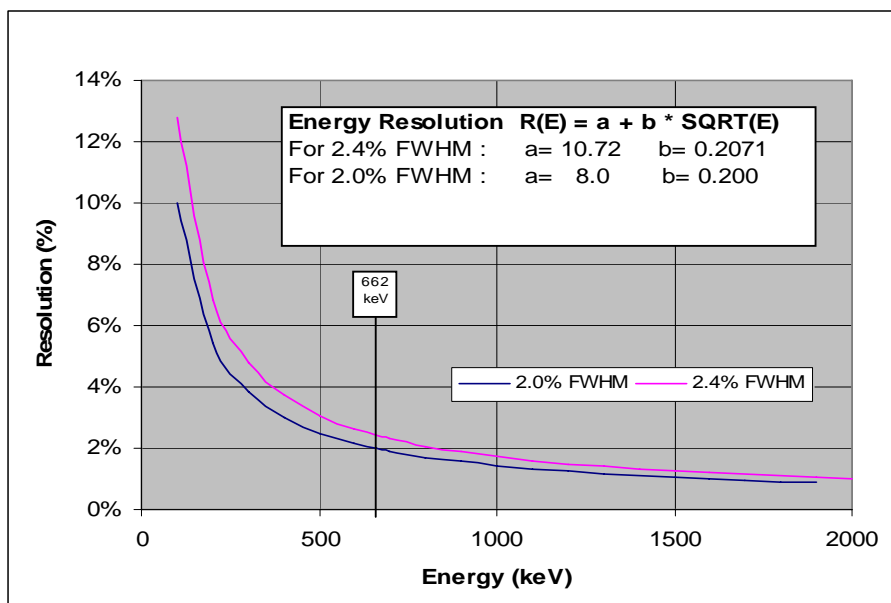


Figure 2 HPXe Test Detector Resolution Calibration

The 9-radionuclide source was then used to measure absolute photopeak efficiency over the 88-1836 keV range for the HPXe detector, as well as for a 30% germanium detector and a 3X3 NaI detector in the same geometry. Figure 3 shows that the photopeak efficiency of the selected HPXe detector (green line/yellow measured points) excels below about 400keV as compared to the 30% germanium detector, and offers similar efficiency as that of the germanium detector above 400 keV. As compared to a 3X3 NaI detector, the test HPXe detector efficiency is greater than that of a 3X3 NaI detector below 200keV, but significantly less at all energies above 200 keV.

Detector Stability

Detector stability with respect to time and temperature is a very important consideration for nuclear monitoring at remote stations and for field measurements with portable radionuclide measurement systems. Spectra with gain shift due to drift with time or temperature are much more likely to result in incorrect identification of radionuclides, particularly if the spectra are transmitted to central stations for analysis.

Measurements previously reported by one of the investigators confirmed that the gain and temperature stability of HPXe detectors is excellent (Beyerle, 2005). Time variance of the gain was measured to be less than $\pm 0.02\%$ per week. The temperature stability of the HPXe detector was measured to be about $\pm 0.2\%$ over the extended range of 25°C–80°C (77°F–176°F). Temperature stability is important for applications such as outdoor air monitoring or vehicle portal monitoring systems where the detector temperature may change significantly on a daily and seasonal basis.

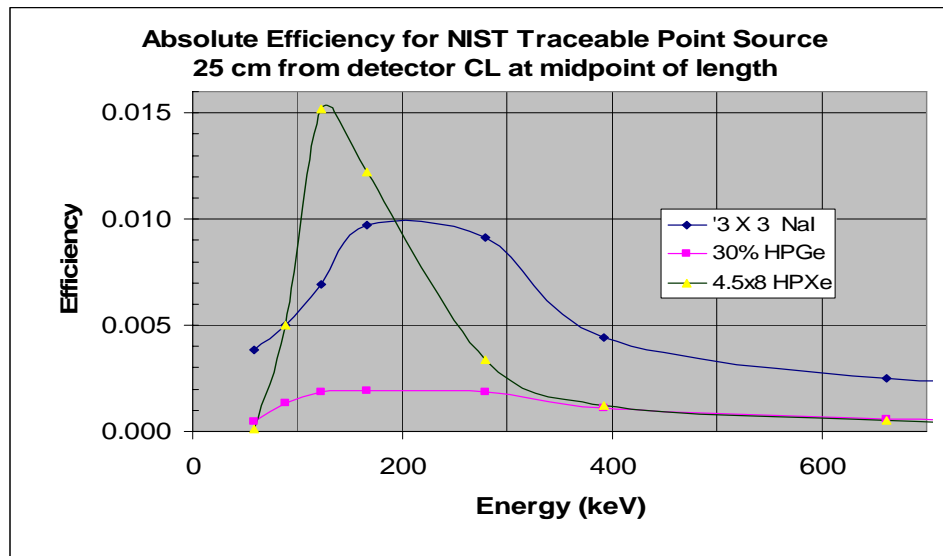


Figure 3 Efficiency Comparison of HPXe, NaI and HPGe

Monte Carlo Simulations

Because Monte Carlo simulations with MCNP5 were used to predict HPXe detector performance not conveniently confirmed by direct measurements in the Phase I project, the ability of the MCNP simulations to accurately predict HPXe performance was experimentally confirmed. Individual HPXe spectra were obtained with commercial sources of ^{57}Co (122.1 keV) and ^{137}Cs (662 keV), and with custom short lived sources of ^{128}I (442.9 keV), ^{64}Cu (511 keV), ^{41}Ar (1293.5 keV) and ^{28}Al (1779.0 keV) prepared and counted at the UC-Irvine research reactor.

Figure 4 shows comparisons over two energy ranges of experimentally measured spectra and MCNP spectra corrected to give the 2.4 % FWHM resolution of the test HPXe detector. Note that all basic features of the photopeak, Compton edge, back scatter edge, escape peaks, and x-ray emissions and edges that appear in the measured spectrum are also apparent in the MCNP simulation.

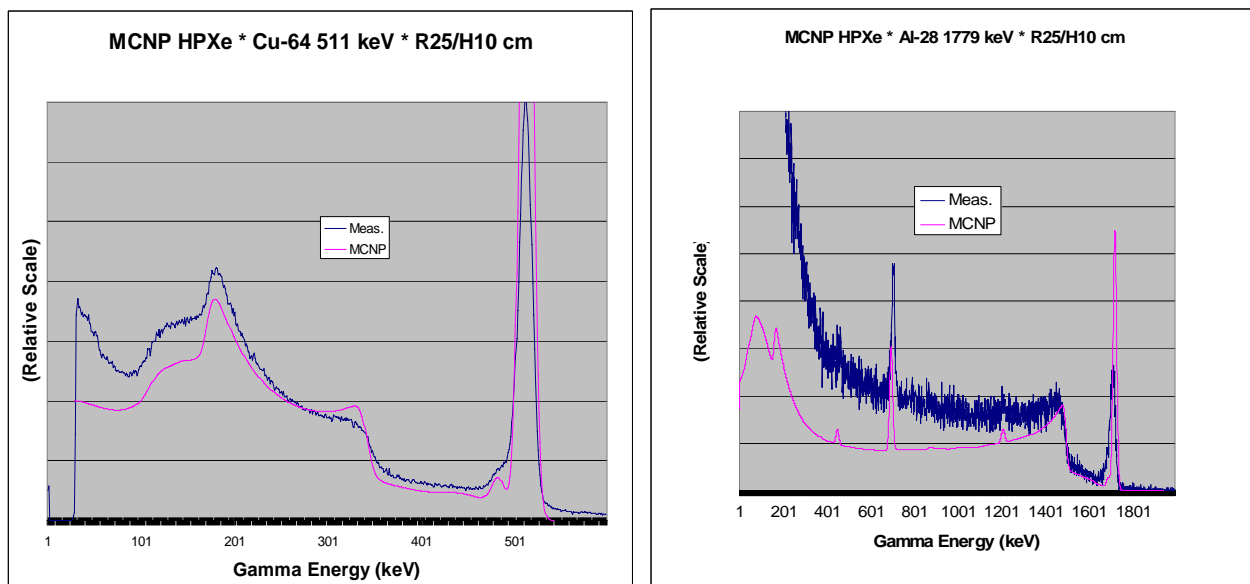


Figure 4 Comparison of Measured and MCNP5 Single Line Spectra for HPXe

The experimental detector runs were made with an unshielded detector, thus giving a much larger background continuum over the low energy range than did the MCNP simulated spectra that did not include background components. A rudimentary shield of lead pieces was constructed around the HPXe detector for an experimental background measurement. The integrated count of the background over the energy range of 100keV -1000 keV for shielded versus unshielded showed a reduction by a factor of more than 14 in gross background counts. The custom designed detector, electronics and shield of the Phase II program will significantly reduce the background, and are expected to bring the MCNP simulation and observed performance into close quantitative agreement.

Once the general credibility and bounds of applicability of the MCNP model were established during Phase I, the model was used to resolve questions and to guide the development effort in several areas:

- A source sample geometry of a concentric cylindrical filter was modeled in MCNP with a 9-radionuclide mix evenly distributed over the cylindrical shell to provide energy-dependent efficiency coefficients for the anticipated concentric cylindrical filter geometry of the NEM prototype system.
- MCNP simulated background spectra were generated for the HPXe detector and source geometry as expected from background components collected during a typical 24 hour NEM sampling period. The input source term for the MCNP runs was taken from concentration levels provided by PNNL from routine sampling runs with the RASA system.
- The effect of xenon gas pressure (xenon density) on detector performance was evaluated as a means to quantify the effect of xenon density on both the efficiency and peak-Compton ratio for a fixed detector volume.
- The effect of the thickness of the steel wall of the HPXe detector was evaluated, as was the improvement in efficiency by constructing the detector wall of aluminum rather than steel. The MCNP results predict that aluminum gives an efficiency improvement for this detector of about 50% at 122 keV, declining to about a 10% improvement at 1836 keV.
- An MCNP model of a 5 cm thick lead shield closely spaced around the cylindrical sample was used to predict the amount of backscatter and pair production in the shield volume from the known aerosol background components in the filter and to ensure that they would not cause interference at crucial points of the gamma spectrum. This model will be used to evaluate the benefit of a copper liner on the inside wall of the shield for the NEM Prototype system of Phase II.

Estimating HPXe Performance for NEM Applications

The second major objective of Phase 1 was to estimate the performance of HPXe detectors for the radionuclide mix of the Comprehensive Nuclear-Test-Ban Treaty-IMS application as defined by previous RASA/Automated Radioxenon Sampler/Analyzer (ARSA) work. In the absence of an air sample collection system and an effective shield for the HPXe test for the Phase I effort, field data from the RASA program provided by PNNL were used to estimate system background and interference under typical IMS sampling conditions.

Performance targets for NEM radionuclide measurement systems were taken from two sources: the NNSA specification of the solicitation, and the IMS specification for radionuclide monitoring systems for certified IMS monitoring stations. The nominal IMS collection/counting cycle is a 24 hour collection period, a 24-hour delay for decay of natural background components on the filter, and a 24 hour counting time.

For IMS certification, NEM radionuclide measurement systems are required to have a detection sensitivity of $\leq 30 \mu\text{Bq/m}^3$ of ^{140}Ba for the nominal counting cycle with collection of a $12,000\text{m}^3$ sample on the filter. The NNSA solicitation for this topic area stated a slightly different requirement of $20 \mu\text{Bq/m}^3$ of ^{140}Ba in air for a $25,000 \text{ m}^3$ sample collected during the 24 hour sampling period. The two specifications are similar in the required detector performance, with the IMS specification requiring slightly better detector performance than the NNSA requirement.

Estimating counts were recorded in HPXe spectra for various concentrations of ^{140}Ba . The five gamma energy emissions from ^{140}Ba that have greater than 1% relative abundance are listed in Table 2. MCNP runs were made with input parameters corresponding to the selected HPXe detector with ^{140}Ba distributed randomly on a concentric cylindrical filter source of 15.2 cm diameter by 20.3 cm length (6 in. diameter by 8 in. length). The IMS cycle of 24-hour collection, 24-hour decay, and 24-hour count time was applied for four cases of airborne ^{140}Ba concentrations and total volumes of air sampled through a used filter. (A "used" filter is defined as a filter containing the airborne

particulates routinely collected by sampling the specified total volume of air at the required collection efficiency, but not containing fresh fission products from a nuclear explosion.)

Table 2 MCNP Calculated Net Counts for Four Test Cases

¹⁴⁰ Ba Decay Data		Counts in ¹⁴⁰ Ba peaks for 4 Sample Cases			
E _γ (keV) f or ¹⁴⁰ Ba Lines of < 1% I _γ	I _γ (%)	20uBq/m3 12,000 m3	30uBq/m3 12,000 m3	20uBq/m3 25,000 m3	30uBq/m3 25,000 m3
162.66	6.2	1498	2110	3340	5028
304.85	4.3	537	768	1154	1761
423.73	3.2	186	274	457	600
437.58	1.9	145	205	305	399
537.26	24.4	947	1379	2068	3065

Estimating Spectrum Baseline and Interference under NEM System Conditions - An analysis of data from the RASA and ARSA programs shows that the major contributors to the natural gamma spectrum baseline for a “used” filter are ⁷Be, ⁴⁰K, ²¹²Pb / ²¹²Bi / ²⁰⁸Tl from the thorium decay series, and ²¹⁴Pb / ²¹⁴Bi from the ²³⁸U decay series (Arthur et al., 2001). During the Phase I project, PNNL provided used RASA filters for counting on the HPXe detector, but the delay caused by logistics of handling and shipping the sample caused the measurements to be unrepresentative because of the rapid decay of the major natural products on the filter.

An analysis of PNNL data from the RASA program indicates that most of the significant gamma peaks and continuum counts in a spectrum for IMS measurement conditions are produced by the gamma emissions from the filter as shown in Table 3. The gamma emission rates of Table 3 were used as input source terms for runs with the MCNP model for the concentric filter around the 2.4% HPXe detector.

Table 3 Gamma Emissions from Used Filter with 24 Hour delay

Gamma Energy (keV)	Intensity %	Gammas/ Second	Radionuclide Source	Gamma Energy (keV)	Intensity %	Gammas/ Second	Radionuclide Source
238.6	43.3	17.41	Pb-212	727.3	6.58	2.87	Bi-212
277.4	6.13	0.94	Tl-208	785.4	1.102	0.48	Bi-212
300.1	3.28	1.32	Pb-212	860.6	12.42	1.91	Tl-208
477.6	10.52	3.34	Be-7	1460.8	11.00	0.78	K-40
510.8	22.6	3.48	Tl-208	1620.5	1.49	0.65	Bi-212
583.2	84.5	13.01	Tl-208	2614.53	99.0	15.25	Tl-208

In order to simulate a NEM spectrum, a composite spectrum was generated by a channel-by-channel summing of counts in the MCNP generated ¹⁴⁰Ba spectrum with counts in the used filter spectrum from MCNP runs for an assumed HPXe resolution of 2.4%. Figure 5 shows both the baseline-only spectrum (blue line) from used filter

background activity, and the composite spectrum of the sum of used filter and ^{140}Ba counts (red line) for the NNSA and IMS specification for NEM systems. The ^{140}Ba peaks at 305, 424 and 438 keV have low intensity and interferences that would prevent their use for ^{140}Ba quantification. The 163 and 537 keV peaks appear to be viable candidates for ^{140}Ba analysis.

The 163 peak will generate the most counts because of the very high efficiency of the HPXe detector in the 100–200keV range. Although the MCNP calculated baseline is fairly low in this energy region, the actual baseline will be larger than shown because of counts from low energy background sources not included in the input source data for the MCNP model.

The 537 keV peak has very good potential to provide quantitative results for the stated ^{140}Ba concentrations and sampling conditions with the 2.4% FWHM HPXe detector. The 537 peak of ^{140}Ba overlaps the 511 peak and x-ray escape peak of the 583 line, but the multiplet resolution routines of commercial analysis software package could readily extract a quantitative number from the data shown. Since the energies of the ^{140}Ba and ^{208}Tl peaks are known, a deconvolution algorithm based on fitting the three Gaussian peaks to the known energies should give even better quantitative results for ^{140}Ba .

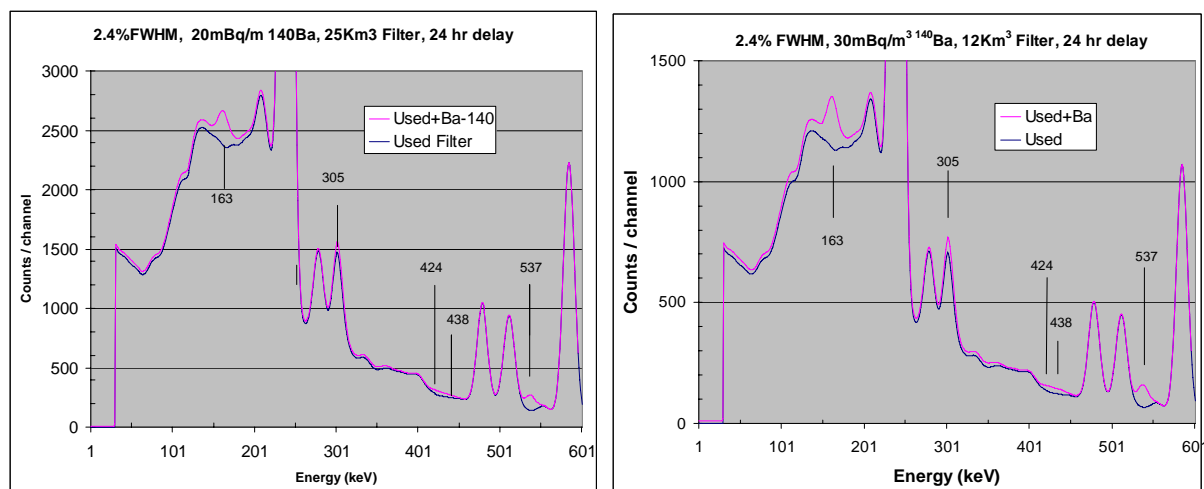


Figure 5 HPXe (2.4% FWHM) Spectra for NNSA and IMS Specifications

One of the first tasks of the Phase II program is to produce an HPXe detector with the same active volume dimensions as the test HPXe detector of Phase I, but with energy resolution of $\leq 2.0\%$ FWHM at 662 keV. A smaller HPXe detector is routinely produced with $\leq 1.7\%$ FWHM resolution, thus, production of the 2.0% detector for the NEM application is expected to be a straightforward engineering development effort. The theoretical resolution limit for HPXe detectors is less than 1%.

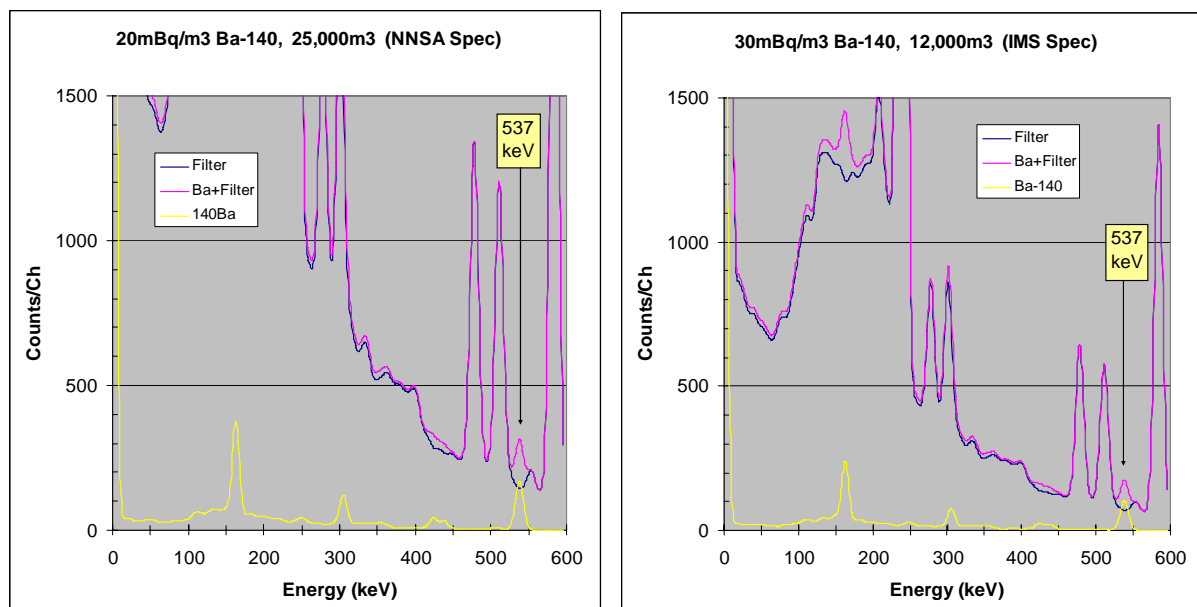


Figure 7 HPXe (2.0% FWHM) Spectra for NNSA and IMS Specifications

Figure 7 shows the calculated spectra for the NNSA and IMS standard conditions with an HPXe detector of 2.0% resolution. The 537 keV ^{140}Ba line is now clearly resolved from the ^{208}Tl peak at 511 keV and the x-ray escape peak of the 538 keV gamma emission of ^{208}Tl .

Estimating Detection Sensitivity - Because the background from the filter could not be experimentally measured on an HPXe detector during the Phase I program, the calculation of the lower detection limit for ^{140}Ba for a HPXe-based NEM system is subject to large uncertainties with the present information. However, a preliminary estimate of detection sensitivity was made using the Currie methods (Currie, 1984) for calculating concentration detection limits at 95% confidence levels for gamma spectra. Input parameters used were the calculated absolute efficiency of 0.015 at 537 keV for the concentric cylindrical filter, and baseline count data in a 13keV window under the 537 keV peak as shown in Figure 7. Using the IMS conditions of a $12,000\text{m}^3$ sample, a 24 hour decay time and a 24-hour counting time, the calculated critical count level is 173 counts, and the concentration detection limit for ^{140}Ba is $11.2\text{ }\mu\text{Bq/m}^3$ for a 2.0% FWHM HPXe detector of the dimensions of the test detector. (The IMS spec is $\leq 30\text{ }\mu\text{Bq/m}^3$.)

Again, the MDC calculation has a very large uncertainty at this stage of the project for the reasons mentioned. A series of bench test measurements with shielded HPXe detectors and freshly collected filter samples will be performed early in the Phase II program to provide the data required to calculate an authoritative MDC for the HPXe NEM Prototype system.

Conceptual Design for the NEM Prototype System

A third goal of the Phase I program was to develop a conceptual design for a Phase II explosion monitor prototype that best meets the specifications and intent of the SBIR subtopic.

An extensive base of practical information was made available to investigators by the developers of the two U.S. HPGe-based NEM systems that have demonstrated the ability to meet the IMS specification. The Investigators were given hosted visits to PNNL in Richland Washington to review an operating RASA monitor, and to DHS-EML in New York City to review an operating AUTORAMP NEM monitor.

The preliminary concept for the NEM Prototype system to be designed and constructed during the Phase II effort is as follows:

Detector—For improved resolution and ruggedness, the NEM HPXe detector will be a gridless design which is expected to give a FWHM resolution of $\leq 2.0\%$ at 662 keV. The dimensions and active detector volume of the NEM Prototype detector will be the same as for the Phase I test HPXe detector, but the detector package size will be

minimized by separating the high voltage power supply and a portion of the signal processing electronics from the detector package.

Filter—A cylindrical pleated filter cartridge of about 14 cm (5.5 in.) I.D. by 20.3 cm (8in.) length, with a nominal 2.5 cm (1.0 in.) thickness will be used in the HPXe-based prototype system. Based on the AUTORAMP pleated filter area multiplication factor of more than 2.5, the effective filter area for the cartridge is about 2430 cm² (377 in²). The cartridge filter concept has proven to be both effective and highly reliable for automated sample changing during field operation of the AUTORAMP system.

Air Sampling System—Air flow for the air sampling system will be provided by a centrifugal blower unit that draws outside air through the filter chamber. A commercially available integral blower and motor unit with continuous duty cycle rating will be purchased that provides the 12,000 flow rate required by the IMS specification. A blower/motor unit similar to the units used in either the RASA or AUTORAMP systems would provide the required airflow performance.

Shield—A custom lead shield will be designed to provide a minimum of 2” thickness of lead. The shield will be a vertical cylindrical design with a split top lid that can be opened and closed under computer control for automated filter cartridge insertion and removal.

System Computer and Control Software—The NEM Prototype system will use an industrial grade PC with a master control program to control operations, provide data acquisition, analysis, archiving, reporting, diagnostics and communications. The master control program will be defined and developed internally as one of the major tasks of the Phase II program, and will integrate commercial off-the-shelf software for spectrum analysis and auxiliary functions where possible to minimize the extent of development of new computer code.

Automatic Filter Cartridge Sample Changer—Since an automatic sample changer is not required to evaluate the detection sensitivity performance of the prototype unit, a sample changer will not be incorporated into the initial NEM Prototype unit. Provisions for adding the sample changing mechanism and software at a later time will be designed into the initial unit based on the requirements of the first field application for the NEM Prototype unit. Commercial robotic XYZ systems of the type used in the AUTORAMP system are available with control software packages that are compatible with operation by the master control program software of the NEM Prototype.

CONCLUSIONS AND RECOMMENDATIONS

The Phase I experimental measurements and MCNP simulations based on previous RASA experiences indicate that a NEM system based on the selected 2.4 % resolution HPXe detector will likely meet IMS and NNSA specifications. Spectra generated and calculated using MCNP5 for ¹⁴⁰Ba and filter baseline show sufficient counts for the 537 keV peak to allow ¹⁴⁰Ba to be detected and quantified at meaningful levels for NEM applications.

The MCNP simulations also indicate that further improvements in detection sensitivity provided by changing to a gridless detector design with $\leq 2.0\%$ energy resolution will further enhance the detection sensitivity for NEM applications. A Phase II proposal has been submitted with detailed goals, work plans and schedules for development and testing of a recommended HPXe-based NEM Prototype system.

During the review of activities in the nuclear non-proliferation area, it became apparent that the NEM program also has an active need for versatile field measurement systems for site inspection and follow up of suspect events. NNSA's need for high resolution ambient temperature portable and transportable nuclear measurement systems is shared by several other US agencies and international groups with nuclear security missions. The rugged, stable HPXe detector appears to have strong potential to provide practical solutions for many of these unfulfilled measurement needs.

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